

Modelling acid-rock interactions and mineral dissolution during RMA stimulation test performed at the Soultz-sous-Forêts EGS site, France

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ABSTRACT

The Soultz EGS reservoir's response to chemical stimulation is assessed by numerical simulation of coupled thermo-hydraulic-chemical processes. To assess chemical interactions between host rocks and a mixture of HCl and HF as well as its potential effects on the Soultz EGS reservoir, new modelling efforts using the FRACHEM code have been initiated. This paper presents the model calibration and results. Simulations consider realistic conditions with available data sets from the EGS system at Soultz. Results indicate that the predicted amount of fracture sealing minerals dissolved by injection of a mixture of acids (RMA) was consistent with the estimated amount from the test performed on GPK4 well at Soultz EGS site. Consequently reservoir porosity and permeability can be enhanced especially near the injection well by acidizing treatment.

1. INTRODUCTION

The Soultz-sous-Forêts Enhanced Geothermal System (EGS), established in the Rhine Graben, north of Strasbourg (France), has been investigated since 1986 (Fig. 1). The final goal of the project is to extract energy for power production from a regional, randomly permeable, natural geothermal reservoir with the complementary resource coming from a forced fluid circulation between injection and production boreholes within a granitic basement. This site was chosen because of the observation of a large thermal anomaly in the region and because of a good knowledge of the shallow geology, due to former oil exploitation in this Tertiary graben. The shallow geology (0 to 1400 m depth) consists of sedimentary layers, overlying the crystalline basement of late-Palaeozoic granites containing hydrothermally altered and fractured zones related to graben normal faults (Genter, 1990; Traineau et al., 1991). It has been observed that deep fluid circulation is supported by the network of permeable fractures. Extensive research has been done to characterize the properties of the fractures. Geophysical borehole measurements including borehole image logs, coring and cuttings analysis showed that nearly-vertical fractures, which show a low permeability, are oriented in an almost North-South direction (Dezayes et al., 1995; Genter et al., 1995). Moreover, it appears that most of the fractures are sealed by hydrothermal deposits, mainly calcite, silica and clays, giving a random distribution to the overall permeability of the system.

After the successful connection to a 3.6 km deep fractured reservoir in 1997, it was decided to look for a new, deeper and hotter reservoir and to build a pilot plant. For this purpose, three wells (GPK3 as a central injection well and GPK2 and GPK4 as production wells) were drilled to 5000 m depth in the crystalline basement to build the EGS system (Fig. 1). The development of Enhanced Geothermal Systems (EGS) depends on the creation of permeable and connected fractures. The Soultz wells have been stimulated hydraulically and chemically in order to develop the underground reservoir (Gérard et al., 2006). The first hydraulic stimulations of the three wells were carried out between 2000 and 2005, and resulted in an improvement of the productivity index of wells GPK2 and GPK4 by a factor of approximately 20 and of GPK3 by a factor of approximately 1.5 (Nami et al., 2007). Although the limited performance of hydraulic stimulation, with high costs and public concern about induced seismic events, provided an important set of reasons for undertaking chemical treatments as additional or even alternative method to hydraulic stimulation, the main argument for chemical stimulation was the evidence, based on drill cuttings and cores analysis as well as on geophysical logs, of fracture filling by carbonates and other soluble minerals.

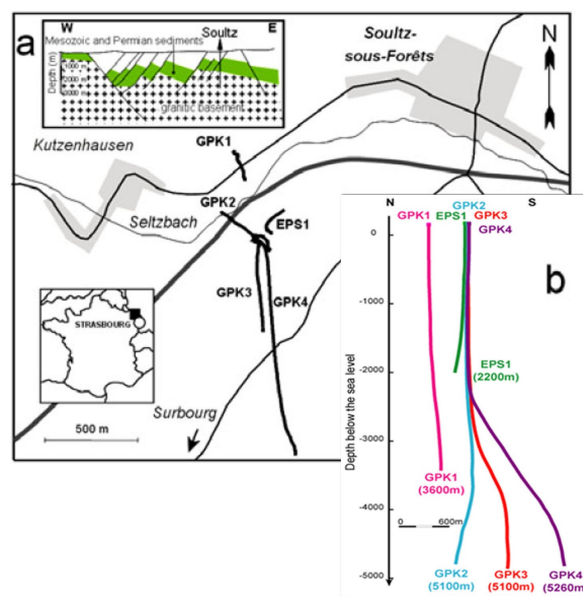


Figure 1. Location of the Soultz-sous-Forêts EGS site (a). Horizontal plan view and schematic geological map of the Rhine Graben (b). N-S cross section and traces of the Soultz deep geothermal wells. Solid lines correspond to well traces (a and b). (After Dezayes et al., 2005).

Removal of secondary mineral filling natural or induced fractures from granitic formation in the near-wellbore vicinity can be accomplished by injecting strong acids (such as HCl and mixtures of HCl-HF). Acid treatments have been successfully applied in many cases to increase or to recover geothermal wells production rates to commercial levels. In order to dissolve the hydrothermal deposits (like carbonates, clay, feldspars and micas) present in the main fracture and porosity zones of the Soultz granite to improve their permeability, Regular Mud Acid (RMA), a mixture of HCl and HF widely used in oil and gas wells, was injected in GPK4 well.

FRACHEM, a thermo-hydraulic-chemical coupled code, was developed especially to forecast the evolution of the EGS project at Soultz-sous-Forêts, Alsace (France). FRACHEM can simulate thermal, hydraulic and fluid-rock interactions within the fractures connecting the injection and the production wells, and determine the dissolution/precipitation reactions of carbonates, pyrite and silicated minerals in the Soultz granite (Durst, 2002; Bächler, 2003; André et al., 2006; Portier et al., 2007). FRACHEM code has been improved to simulate the propagation of reacting fluids and to gain insight into the effectiveness of acidizing treatment as a well stimulation technique. The RMA stimulation method has been applied by numerical modelling to the Soultz EGS system and compared to field observations, to investigate its impact and effectiveness.

2. FIELD TEST: STIMULATION OF GPK4 WITH REGULAR MUD ACID

Hydrofluoric acid (HF) is the only common acid that dissolves clay, feldspar and quartz fines. For years mixtures of HF and HCl (RMA treatment) have been the standard acidizing treatment to dissolve these minerals that cause damage. In sandstone acidizing treatments, a preflush of HCl varying between 7.5% to 15% is usually injected ahead of the HCl/HF mixture to dissolve the carbonate minerals and avoid precipitation of calcium fluoride. The minimum volume is determined by assuming that the HCl-carbonate reaction is very fast so that the HCl reaction front is sharp.

Regular Mud Acid (RMA) was injected from the wellhead through the casing string in GPK4 well. The stimulation zones were therefore the whole openhole section of the well (500 to 650 m length).

In May 2006, the RMA treatment was carried out in four steps with addition of a corrosion inhibitor when needed. Before the injection of RMA, 2000 m³ of cold deoxygenated water were introduced in the well at 12 L.s⁻¹, then at 22 L.s⁻¹, and finally at 28 L.s⁻¹. Later, to avoid calcium fluoride (CaF₂) precipitation that can lead to well damage, a preflush of 25 m³ of a 15% solution of HCl in deoxygenated water (3.75 tons of HCl) was pumped ahead of the HCl-HF acid mixture for 15 minutes at 22 L.s⁻¹. A main flush consisting of a total of 200 m³ of 12/3 (wt%) Regular Mud Acid (RMA) was then injected at a flow rate of 25 L.s⁻¹ for 2.5 hours. Finally, a postflush of 2000 m³ of cold deoxygenated water, at a flow rate of 22 L.s⁻¹, then 28 L.s⁻¹ during 1 day, was injected after the RMA injection.

After the RMA stimulation, the wellhead pressure curve was smooth, indicating an efficient clean-up of the hydrothermalized fracture or porous zones in the first ten meters surrounding the open hole (GEIE, 2006) (Fig. 2). The step rate test performed later in May 2006 (Fig. 2),

after the RMA-stimulation, shows that, after three days of injection, the wellhead pressure is about 65 bars, which is about 16 bars lower than before stimulation (step rate test performed on April 2006). This represents a 35% reduction of the wellhead pressure due to acidification treatment (GEIE, 2006). Before the RMA treatment, the wellhead pressure curve raised with the flow increase showing a restricted storage capacity in the vicinity of the well. It can be estimated that the RMA-stimulation has therefore resulted in a maximum enhancement of the injectivity index of 35%. However, no production test was performed after these operations to verify the productivity index.

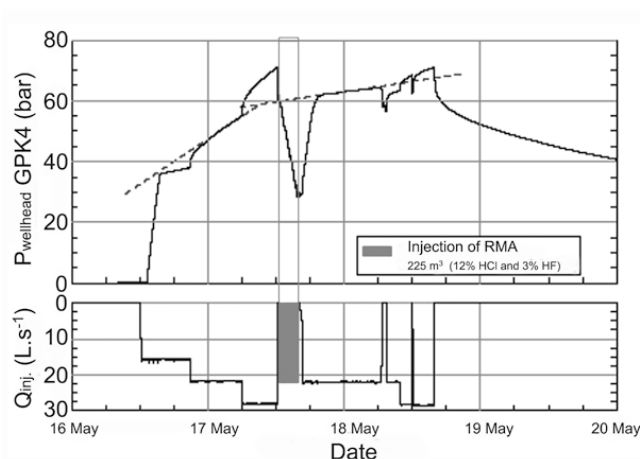


Figure 2. Impact of Regular Mud Acid acidification test performed on GPK4 in May 2006 (after Nami et al, 2007). The slope change of the pressure (dash line) for the same flow rate before and after RMA injection (preflush of 25 m³ HCl at 15% and mainflush of 200 m³ RMA, at 22 L/s during ~3 hours) indicates a gain in productivity. (After Nami et al., 2007)

3. MODEL SETUP

The mineral dissolution and porosity enhancement near the injection well following short acid mixture injection has been reproduced by simulation using FRACHEM.

3.1 Geometrical and flow conditions

The present application of FRACHEM is the modelling of a 2-D simplified model with a geometry close to the Soultz system. Injection and production wells are linked by fractured zones and surrounded by the impermeable granite matrix. The model is composed of 1250 fractured zones. Each fractured zone has an aperture of 0.1 m, a depth of 10 m, a porosity of 10%, and contains 200 fractures. This model allows an effective open thickness of about 125 m, while the mean openhole section of each well is about 600 m. Initially the temperature was set to the reservoir temperature of 200°C and the fractured zone contains the formation fluid.

One of these fractured zones is modelled with the assumption that the fluid exchange with the surrounding low permeability matrix is insignificant. Due to the symmetrical shape of the model, only the upper part of the fractured zone is considered in the simulation. The area is discretized into 222 2D elements (Figure 3).

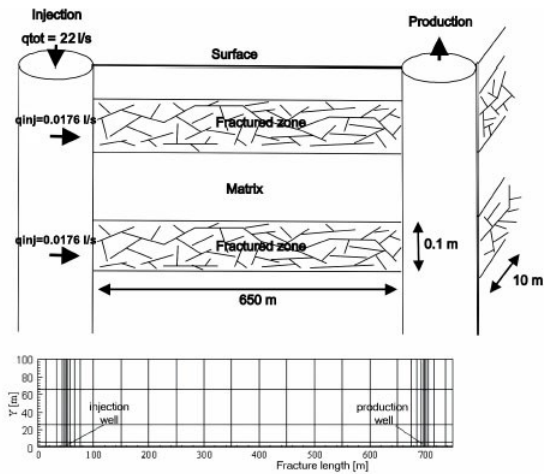


Figure 3. Simplified model and spatial discretization.

Considering the main flush injection rate of 22 L/s, the fluid was re-injected in the modelled fractured zone at a rate of $1.76 \cdot 10^{-2}$ L/s. During this simulation a constant overpressure of 70 bars was assumed at the injection well and an initial hydrostatic pressure of 500 bars was assumed for about 5000 m depth. The uncertainty on the permeability specification doesn't affect modelling results of reactive transport and porosity enhancement because a constant injection rate was specified. Dirichlet boundary conditions were applied to the upper, left and right side of the model. The values of thermo-hydraulic parameters considered in the simulation are listed in Table 1.

An injection temperature of 65°C was used. Injection water chemistry was the same as in the field test, a 3wt% HF-12wt% HCl solution, and a density of 1075 kg/m³. The initial water chemistry is in equilibrium with the initial mineralogy at a reservoir temperature of 200°C. A maximum test-period of two days was simulated, including injection of cold fresh water at 25°C, a preflush of 15wt% HCl solution during 0.25 hours and a postflush period of one day after the 2.5 hours injection of RMA.

3.2 Mineralogical and chemical settings

The mineralogical composition of Soultz granite given by Jacquot (2000) on GPK2 is assumed to be the same for the three wells (GPK2, GPK3 and GPK4) (Table 2). In the following simulation, the fluids are assumed to circulate within the hydrothermalised granite. Concerning the mineralogical composition of the fractured zone, only quartz (40.9 %), K-feldspar (13.9 %), calcite (3.3 %), dolomite (0.8 %) and illite (24.6 %) are considered.

The geothermal fluid present in the formation is a NaCl brine with a pH of 4.9, a total dissolved solids of about 100 g/l and a temperature at the beginning of the simulation of 200 °C. The main characteristics of this fluid are given in Table 3.

Table 1. Thermo-hydraulic parameters for the main flush

Reservoir properties			
Parameters	Fracture	Matrix	Fluid
Hydraulic conductivity [m ² /Pa.s]	$7.4 \cdot 10^{-8}$	10^{-15}	-
Thermal conductivity [W/m.K]	2.9	3	0.6
Density [kg/m ³]	-	2650	1000
Heat capacity [J/kg.K]	-	1000	4200
Porosity [%]	10	0	-
Initial and boundary conditions			
Injection Overpressure (bar)	70		
Temperature (°C)	200		
Injection conditions			
Temperature (°C)	65		
Rate (L/s)	22		
Duration (hours)	2.5		

Table 2. Mean composition (in volume percent) of the different facies of granite in the Soultz reservoir (Jacquot, 2000).

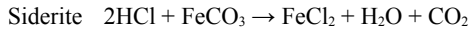
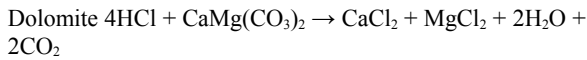
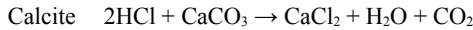
Minerals	Healthy granite	Hydrothermalised granite	Vein of alteration
Quartz	24.2	40.9	43.9
K-Feldspar	23.6	13.9	
Plagioclases	42.5		
Illite		24.6	40.2
Smectite		9.7	9.6
Micas	9.3		
Calcite	0.3	3.3	4.3
Dolomite		0.8	0.7
Pyrite		0.7	1.0
Galena		1.3	0.3
Chlorite		4.8	

Table 3. Characteristics of the geothermal fluid used for the numerical simulation

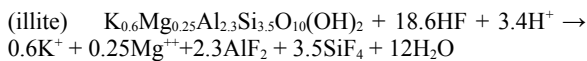
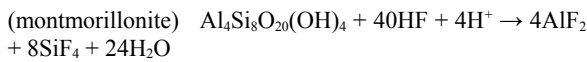
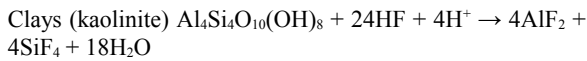
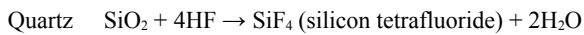
Fluid		Formation brine
Temperature (°C)		200
pH		4.9
Concentration (mg/kg)	Na ⁺	26400
	K ⁺	2870
	Ca ²⁺	6160
	Mg ²⁺	112
	Fe ²⁺	134
	SiO ₂	364
	Cl ⁻	54205
	SO ₄ ²⁻	63
HCO ₃ ⁻	58	

3.3 Dissolving power of acids

The main readily HCl-soluble minerals are calcite, dolomite, and siderite which additionally do not generate precipitates. The reactions are:



Siliceous minerals are dissolved by hydrofluoric acid and its chemistry is much more complex than the HCl when reacting with carbonates (Walsh et al., 1982; Pournik, 2004). Quartz, clay, and feldspars are the main siliceous particles involved in damage of sandstones. The primary chemical reactions in sandstone acidizing are:



A convenient way to express reaction stoichiometry is with the dissolving power, introduced by Williams et al. (1979). The dissolving power expresses the amount of mineral that can be consumed by a given amount of acid on a mass or volume basis. X_g , the mass of mineral consumed by a given mass of acid, is defined as:

$$X_g = \frac{V_{\min \text{ eral}} M_{W, \min \text{ eral}}}{V_{\text{acid}} M_{w, \text{acid}}}$$

The dissolving power of any concentration of acid is the X_{g100} times the weight fraction of acid in the acid solution. For the commonly used preflush of 15 wt% HCl, $X_{g15} = 0.15(X_{g100})$. The stoichiometric coefficients for common acidizing reactions are found from the reaction equations described above, while the molecular weights of the acids and minerals considered in the simulation are listed in Table 4.

The volumetric dissolving power, X_v , similarly defined as the volume of mineral dissolved by a given volume of acid, is related to the dissolving power X_g by:

$$X_v = X_g \frac{\rho_{\text{acid solution}}}{\rho_{\min \text{ eral}}}$$

The volumetric dissolving power of HCl with calcite and dolomite and for HF with quartz and albite are given in Table 5.

The volume of HCl preflush needed to consume the calcite to a short distance from the wellbore is the volume of calcite present divided by the volumetric dissolving power.

The main acid stage requires the greatest emphasis because of the damage mechanisms, directly associated to precipitation of products from the HF reactions (Allen and Roberts, 1989; Kalfayan, 2001). Acid stimulation techniques have to account for both chemistry and treatment execution to accurately predict the effectiveness since the effect of these precipitates could be minimized if they are deposited far from the wellbore (Entingh, 1999). Careful selection of mixtures, additives, acids formulations, and treatment volumes must be accounted to minimize these secondary adverse effects.

Table 4. Molecular weights and density of the acids and minerals considered in the simulation

Compounds	Molecular weight (g/mol)	Density (g/cm ³)
HCl	63.5	15%wt HCl solution: 1.07
HF	20	3%wt HF-12%wt HCl solution: 1.075
Calcite	100.1	2.71
Dolomite	184.4	2.84
Siderite	115.8	3.94
Quartz	60.1	2.65
Albite	262.3	2.61
K-feldspars	278.4	2.5
Illite	389.3	2.75
Am.Silica	60.1	2.07

Table 5. Volumetric dissolving power of RMA

Mineral	Acid concentration 12%wt HCl - 3%wt HF
Calcite	0.067
Dolomite	0.058
Quartz	0.010
K-feldspar	0.011

3.4 Acid mineral reaction kinetics

Reaction rates are affected for kinetics; among the factors that strongly influence the mineral reactions are acid concentration and temperature. Dissolution reaction rates are proportional to the HF concentration for most sandstone minerals. The dissolution of minerals is a thermally activated phenomenon; thus, the rates increase greatly as a function of temperature, and the penetration depths of live acid diminish accordingly. Reaction kinetics data have been found in the literature for the reactions of HCl with calcite and dolomite and for the reactions of HF with quartz, feldspars and clays. Efforts have been made to develop FRACHEM code to simulate the acidizing process.

Acid-mineral reactions are termed heterogeneous reactions because they are reactions between species occurring at the interface between different phases, the aqueous phase acid and the solid mineral. The kinetics of a reaction is a description of the rate at which the chemical reaction takes place, once the reacting species have been brought into contact.

The reaction rate for the HCl-CaCO₃ reaction is extremely high, so the overall rate of this reaction is usually controlled

by the rate of acid transport to the surface. On the other hand, the surface reaction rates for many HF-mineral reactions are very slow compared with the acid transport rate, and the overall rate of acid consumption or mineral dissolution is reaction rate controlled.

Reactions of HCl with carbonates

HCl is a strong acid, meaning that when HCl is dissolved in water, the acid molecules almost completely dissociate to form hydrogen ions, H^+ , and chloride ions, Cl^- . The reaction between HCl and carbonate minerals is actually a reaction of the H^+ with the mineral. Lund et al. (1973, 1975) measured the kinetics of the HCl-calcite and HCl-dolomite reactions, respectively. Their results were summarized by Schechter (1992) as follows:

$$-r_{carbonate} = sE_f (C_{HCl})^\alpha$$

$$E_f = E_f^0 \exp\left(\frac{\Delta E}{RT}\right)$$

The constants are given in Table 6. s is the surface area of the mineral (in m^2). SI units are used in these expressions, so C_{HCl} has units of mole/ m^3 and T is in K.

Reaction of HF with sandstone minerals

HF reacts with virtually all of the many mineral constituents of sandstone. Reaction kinetics have been reported for the reactions of HF with quartz (Bergman,

1963; Hill et al., 1981), feldspars (Fogler et al., 1975), and clays (Kline and Fogler, 1981). These kinetic expressions can all be represented by:

$$-r_{mineral} = sE_f \left[1 + K(C_{HCl})^\alpha\right] (C_{HF})^\beta$$

and the constants are given in Table 6.

These expressions show that the dependence on HF concentration is approximately first order ($\alpha = 1$). For the feldspar reactions, the reaction rates increase with increasing HCl concentration, even though HCl is not consumed in the reaction. Thus, HCl catalyzes the HF-feldspar reactions. Also, the reaction rates between clay mineral and HF are very similar in magnitude, except for the illite reaction, which is about two orders of magnitude slower than the others.

Per unit mass of rock, the specific surface area of each mineral is its specific surface area times the mass fraction of the mineral present in the sandstone. The fraction of HF expended in a particular reaction is the overall reaction rate for that mineral divided by the sum of the reaction rates. The reaction rates of HF with clays and feldspars are approximately two orders of magnitude higher than that between HF and quartz. Because the clay and feldspars reaction rates are relatively high and they generally comprise a small portion of the total rock mass, they will be consumed first in sandstones acidizing. The quartz reaction becomes important in regions where most of the clay and feldspar have already been dissolved.

Table 6. Kinetic parameters used in the simulation

Mineral	α	β	K	E_f^0	$\Delta E/R$
Calcite	0.63			7.314×10^7	7.55×10^3
Dolomite	$\frac{6.32 \times 10^{-4} T}{1 - 1.92 \times 10^{-3} T}$			4.48×10^5	7.9×10^3
Quartz	-	1	0	2.32×10^{-8}	1150
K-feldspar	0.4	1.2	$5.66 \times 10^{-2} \exp(956/T)$	1.27×10^{-1}	4680
Albite	1	1	$6.24 \times 10^{-2} \exp(554/T)$	9.5×10^{-3}	3930
illite	-	1	0	2.75×10^{-2}	6540

4. SIMULATION RESULTS AND DISCUSSION

To test the above findings, the numerical results were applied to hydraulic data measured during the RMA stimulation test. Since, in the numerical model, the pressure at the injection and the production point was fixed, pressure cannot be used for comparison. Instead, the transmissivity evolution was compared.

Figure 2 shows the pressure evolution at GPK4 (injection well) during the stimulation test as well as the fluid injection rate.

The transmissivity, T , is calculated by:

$$T = \frac{q_{tot}}{L \|\nabla P\|}$$

q_{tot} is the fluid injection rate, L the depth of the fracture and ∇P the pressure gradient. In the case of the numerical model, L is 10 m, ∇P is constant, since the pressure was

fixed at the injection and the production point. q_{tot} is calculated by:

$$q_{tot} = v_f A_i \Phi$$

v_f is the fluid velocity, A_i the area of the element interfaces and Φ the porosity. The resulting flow rate evolution in the fractured zone is illustrated in Figure 4.

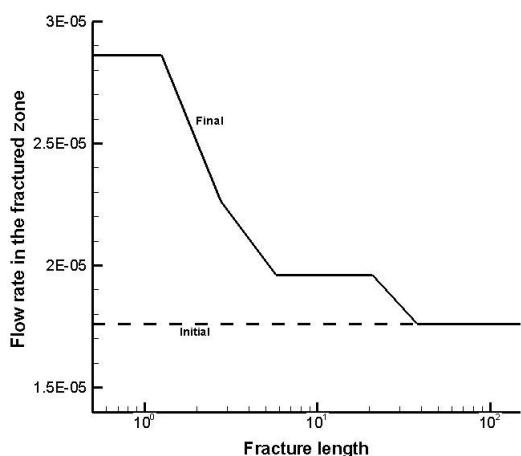


Figure 4. Resulting flow rate (in m^3/s) evolution as a function of the fractured zone length (in m) from numerical model.

To calculate the transmissivity evolution during the stimulation test according to the numerical model, that is for a single fractured zone, the fluid injection rate is divided by the number of assumed fractured zones. ∇P is taken from the stimulation data. Due to different initial pressure conditions, the transmissivity in the numerical model is higher than that in the stimulation test. However, the transmissivity evolution as a percentage of the initial value in the first meters of the fractured zone increases in both cases by 35%. The fact that the transmissivity in the stimulation test increases points to geochemical processes in the reservoir, such as the dissolution of calcite and feldspars. The good fit between the transmissivity evolution of the numerical model and the stimulation test confirms this assumption. The pressure evolution in the reservoir is therefore the result of geochemical processes.

Overall enhancement of porosity obtained from the simulation is presented in Figure 5. The porosity increases to about 0.17 from an initial value of 0.1 close to the injection point. The enhancement of porosity extends to a distance of about 30 m. Increases in porosity are mainly caused by dissolution of calcite, K-Feldspar, albite and illite (Figure 6). Results show that acid preflush dissolves carbonates in the first metres of the fractured zone. Calcite dissolves with maximum amount of 3.3% in the first 1,5 metres from the injection point. The other minerals are not attacked by the 15%wt HCl solution. When injecting the RMA treatment, the influenced zone extends and the first 40 metres around the injection well are affected (Figure 6). Because the clay and feldspars reaction rates are relatively high and they comprise a small portion of the total rock mass, they are consumed first in RMA acidizing. K-feldspar, illite and albite dissolution occurs close to the injection point (Figure 6). The quartz reaction becomes important in regions where most of illite and feldspars have already been dissolved. However, amounts of quartz dissolution are very small because of lower reaction rate.

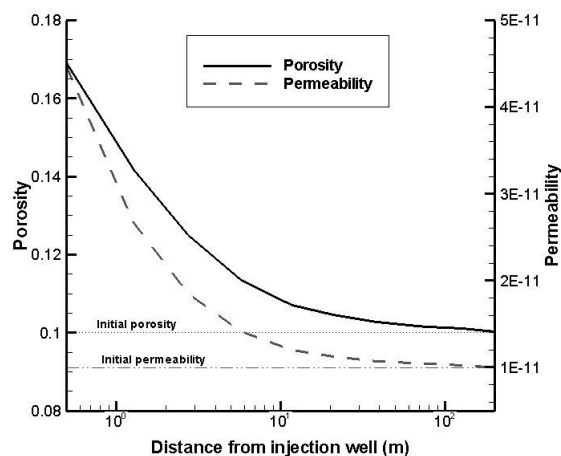


Figure 5. Distribution of porosity and permeability (in m^2) enhancement obtained from the simulation.

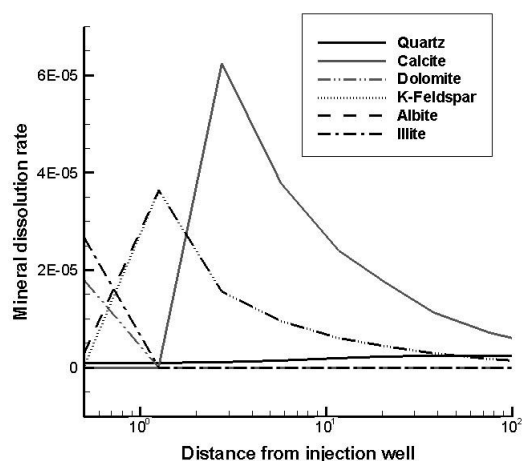


Figure 6. Evolution of the mineral dissolution rates (in $\text{mol}/\text{m}^3/\text{s}$) with distance (in m) following RMA injection.

5. CONCLUSIONS

Recently, sandstone acidizing treatments have been successfully performed in geothermal granitic reservoirs such as at Soultz and encouraging results were obtained on GPK4 well.

Some numerical simulations using FRACHEM code were performed to better understand the behaviour of acid mixtures within the reservoir. Considering the geometrical model used for the simulations and the different assumptions about the fluid and rock compositions, some estimation has been proposed.

The injection of a regular mud acid (RMA) solution results in dissolution of calcite, clays and feldspar minerals. Consequently reservoir porosity and permeability can be enhanced in a region extending several meters around the injection well. Nevertheless, the high reactivity and a weak flow prevent the penetration of acid in the far field between the wells. This high reactivity also involves the risk of

creating wormholes, able to increase the porosity but not always the permeability of the fractured reservoir.

Many factors affect mineral dissolution and associated enhancement in formation porosity and permeability, including mineral abundance and distribution in the formation, reaction kinetics, and injection rate. More detailed investigations will be conducted in the future.

Finally, this study demonstrated that acid mixtures injection can play a significant role in the development of porosity around injection wells. It was shown by numerical modelling that mixtures of acids have the possibility to react with carbonates and silicates, dissolving them and opening new pores within the reservoir. The positive effect of acid injection on porosity is proportional to the amount of injected acid. Looking for commercial production rates of the wells, other chemical stimulation techniques should be considered such as fracture acidizing. This type of acid job should reach the fracture network of the far field and connect the injection and production wells.

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